Eos, Vol. 86, No. 8, 22 February 2005

EOS

EOS, TRANSACTIONS, AMERICAN GEOPHYSICAL UNION

VOLUME 86 NUMBER 8
22 FEBRUARY 2005
PAGES 77–88

Analyzing Atmospheric Trace Gases and Aerosols Using Passenger Aircraft

PAGES 77, 82–83

CARIBIC (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container) resumed regular measurement flights with an extended scientific payload in December 2004. After an automated measurement container was successfully deployed on intercontinental flights using a Boeing 767 from 1997 to 2002, a far more powerful package now is deployed using a new Airbus A340-600 made available by Lufthansa German Airlines (Star Alliance). The new CARIBIC system will help address a range of current atmospheric science questions during its projected lifetime of 10 years.

European and Japanese scientists are developing a variety of atmospheric chemistry research and monitoring projects based on the use of passenger aircraft. This is a logical approach with a main advantage being that near-global coverage is obtained, in contrast to limited coverage through research aircraft-based expeditions. Moreover, highly detailed and consistent data sets can be acquired, as compared to satellite observations in general. In addition, even compared to land-based observatories, operational costs are moderate.

A key question to ask about these passenger-aircraft based projects is whether the present endeavors are temporary explorations of alternative observation platforms, or whether they are serious systems for monitoring atmospheric composition for decades to come. Are passenger aircraft the "poor man's research aircraft or satellite"? Or, is there a much stronger case to be made for using aircraft for this purpose?

Three presently-operating passenger aircraft systems, including CARIBIC, have been in existence for a considerable period of time, and each one is based on a different concept. A fourth system, noted below, has been terminated.

An Overview of Projects

CARIBIC (www.caribic-atmospheric.com) presently uses one aircraft that one to two times per month carries a full-sized automated "flying laboratory container," integrating a range of analyzers and samplers. The idea is to maximize the number of trace compounds that can be detected and/or sampled for retrospective laboratory analyses. CARIBIC has some flexibility, as the equipment package in the container can be continuously optimized and extended. CARIBIC is evolving to optimize the amount of species that can be detected, although its geographical coverage is less than that obtained by MOZAIC and JAL, which are discussed below.

MOZAIC (Measurement of Ozone and Water Vapour by Airbus In-Service Aircraft; www.aero.obs-mip.fr/mozaic/) has become known because of its ozone and water vapor measurements. The basic idea of having instrument packages for in situ measurement in several aircraft has worked well using five Airbus 340-300 aircraft. In 2001, MOZAIC extended the package in one aircraft to include NO_y measurement, and all five aircraft were fitted with CO analyzers. Presently, MOZAIC, funded by the European Commission (EC), is developing new, compact instrument packages and will be working on aviation certification for A340 series aircraft. MOZAIC is evolving to achieve global coverage for the most important measurements, i.e., O_3, H_2O, CO, and even certain aerosol categories.

The JAL Foundation (www.jal-foundation.or.jp/) supports systematic flights over many years with Japan Airlines to collect air samples in flight using a Boeing 747. The information obtained mainly on routes between Japan and Australia has been useful as an addition to the ground-based station flask sampling by the U.S. National Oceanic and Atmospheric Administration's (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL), Global Atmosphere Watch (GAW), and others.

The Japanese project (a cooperation between the Meteorological Institute of Japan and Japan's National Institute for Environmental Studies) is now expanding to involve seven Boeing 747-400 and 777 aircraft, of which two will perform air sampling and five will be equipped for in situ measurement of CO_2. The JAL project is evolving as a hybrid between the MOZAIC approach of obtaining a large coverage and the approach of using flask sampling (like the NOAA/CMDL network) combined with the in situ observation of CO_2.

NOXAR (Measurements of Nitrogen Oxides and Ozone Along Air Routes; www.iac.ethz.ch/en/research/chemie/tpeter/Noxar.html) was a project to study the effects of aircraft emission of nitrogen oxides on the ozone chemistry of the tropopause region by measuring O_3, NO, and NO_2. Despite the success of NOXAR and the existing lack of systematic data, especially for reactive nitrogen species, the project was terminated.

These civil aircraft-based atmospheric research projects follow a U.S. precursor project GASP (Global Atmospheric Sampling Project) [*Falconer and Holdeman*, 1976; *Pratt and Falconer*, 1979]. Using passenger or freight aircraft to undertake scientific research requires the compatibility of the equipment with the aircraft environment, and certification of the equipment by national and international aviation authorities. These are complex procedures. The interface between analytical science and civil aviation often brings up unexpected questions, but experience has proven that there is a sufficient degree of compatibility.

Although some atmospheric scientists may be skeptical about the need to use passenger (or freight) aircraft, or about the added scientific value, CARIBIC, MOZAIC, and JAL are developing strongly and actually do fill important gaps in atmospheric monitoring left by satellite-based remote sensing of the troposphere, land/ship based operations, and campaign-type research that uses dedicated aircraft. Cautious optimism is warranted by the apparent success of using passenger aircraft for detailed, regular observations of the changing chemistry and composition of the Earth's atmosphere.

Overview of the CARIBIC Program

While each of the civil aircraft-based programs has its own philosophy for development, the common challenge is to carry out measurements and optimize scientific value without

By C. A. M. Brenninkmeijer, F. Slemr, C. Koeppel, D. S. Scharffe, M. Pupek, J. Lelieveld, P. Crutzen, A. Zahn, D. Sprung, H. Fischer, M. Hermann, M. Reichelt, J. Heintzenberg, H. Schlager, H. Ziereis, U. Schumann, B. Dix, U. Platt, R. Ebinghaus, B. Martinsson, P. Ciais, D. Filippi, M. Leuenberger, D. Oram, S. Penkett, P. van Velthoven, and A. Waibel

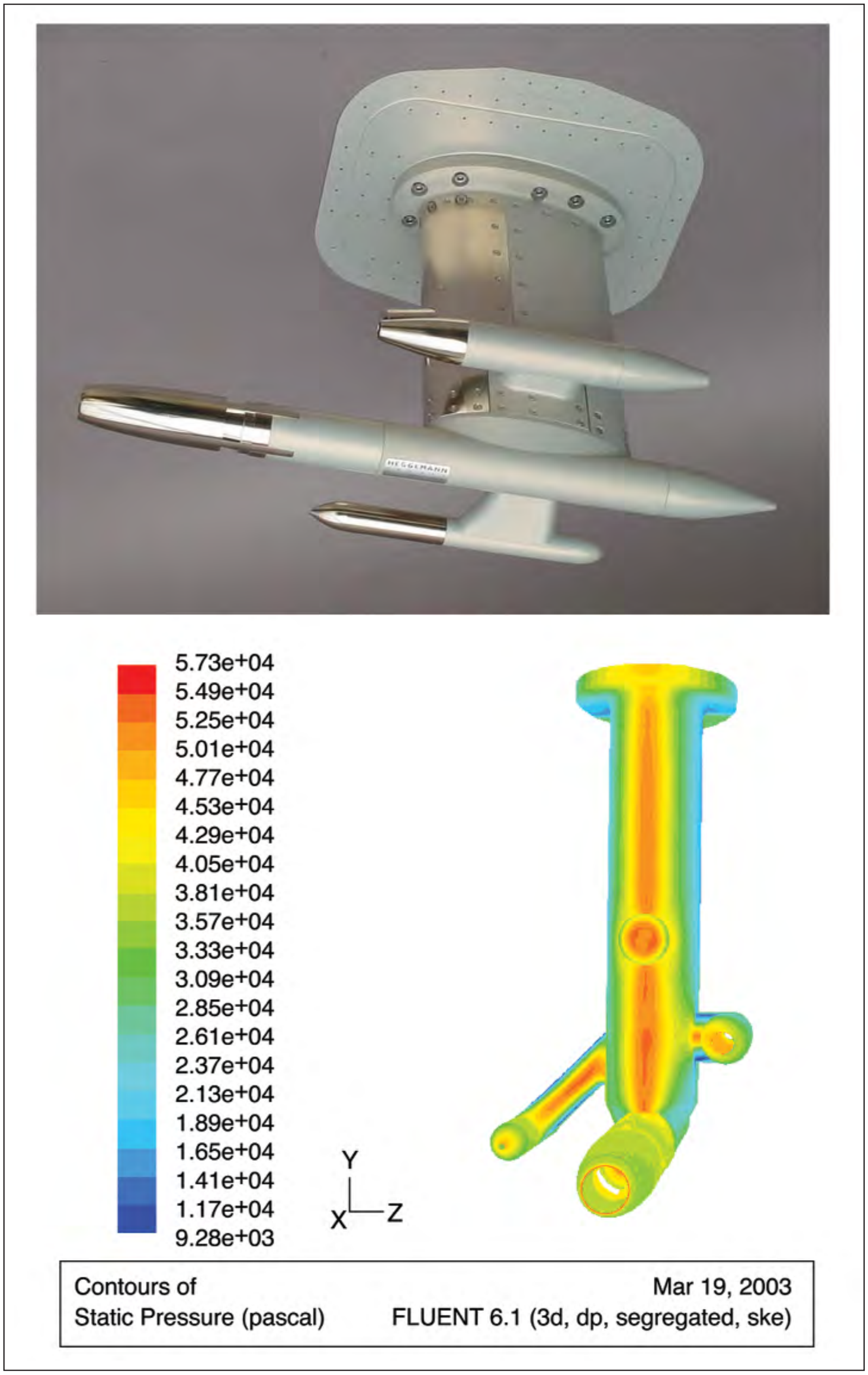

Fig. 1. (top) The inlet system featuring the three individual probes for H_2O, aerosol, and trace gases (left side in flight). The video camera is in the pylon, visible behind the nickel-plated trace gas inlet cone. (bottom) The pressure distribution at the front of the inlet, ranging from about 150 to 550 mbar.

disturbing the complex technical and operational environment of the aircraft.

Within the constraints of being a guest on board a passenger aircraft, the CARIBIC philosophy is to regularly fly and analyze as wide a range of trace compounds as possible. CARIBIC was in operation during 1997–2002 using a Boeing 767-ER (phase 1), and resumed measurements in December 2004, using an Airbus A340-600 (phase 2). For this purpose a new inlet system was developed and the aircraft was modified to accommodate the scientific systems. Also, a new container with new equipment was constructed. Here, the article focuses on the main relevant technical aspects and then highlights some of the existing and anticipated scientific output.

Technical Aspects

The two main components of CARIBIC are the automated instrument container and the air inlet system. There are two fundamental limits to what one can measure with large passenger aircraft. One is that only fully automated equipment compatible with safety requirements can be used. There is no "container operator" on board, and substances like liquid nitrogen cannot be used.

The second limitation concerns the inlet system. This necessarily is of modest dimensions, which rules out the assaying of highly reactive short-lived radical species like OH and RO_x that require large, complex inlet systems. Another category of compounds difficult to measure at cruising speeds of over 250 m/s are aerosol particles.

During phase 1 of CARIBIC, a fairly basic inlet system with one inlet probe for gases and one for aerosol particles functioned well [*Brenninkmeijer et al.*, 1999]. For phase 2, an improved inlet system was designed and constructed. Ideally, the air inlet system should be positioned toward the nose of the aircraft because the thickness of the skin boundary layer increases along the length of the fuselage. However, because of cargo logistical considerations, the CARIBIC container is located in the far end of the forward cargo bay toward the belly fairing, adjacent to the wing box, i.e., about 25 m from the aircraft's nose. In order to keep the sampling lines as short as possible, the air inlet is located nearby (2.5 m away), just in front of the belly fairing.

Figure 1 (top) is a photograph of the new inlet system, with Figure 1 (bottom) showing the calculated pressure distribution around the inlet. Using detailed information supplied by Airbus in Hamburg about the airflow around the fuselage, the distance from the aircraft skin needed to reach air outside the boundary layer has been calculated. The most prominent part of the inlet is the lowermost 60-cm-long aerosol inlet which has a leading shroud. This shroud mainly reduces the effects of changes in the angle of the incoming air on aerosol counts. Inside the 50-cm-long diffuser tube (with a frontal orifice of 6 mm), which has the function of reducing the relative air speed from approximately 250 m/s to several meters per second, is the actual forward-pointing 3/8-inch aerosol intake tube. The overall aerosol sampling efficiency of the phase 1 system was about 70% for ultrafine particles (4–12 nm) and 70–90% for larger particles up to 0.8 μm in diameter. The phase 2 system is expected to have a similar sampling efficiency.

For trace gas measurements, air is taken in through a second diffuser tube, which still retains most of the ram pressure (total pressure of the incoming air) to ensure a high throughput of air through the tubing system connecting the inlet with the container. The third inlet system contains two inlet tubes, one in the nose cone for total water measurement, the other one at the side, for gaseous water only. The inlet tips and tubing are in most instances heated to prevent icing and to prevent loss of trace gases on the walls. One of the parallel trace gas tubes inside the aircraft is heated and lined with perfluoroalkoxy (PFA), and the other tube is electro-polished stainless steel. The water tubes are heated, electro-polished

Fig. 2. Frontal view of the automated measurement container, 3.1 m wide, gross weight, 1450 kg.

stainless steel tubes. The inlet contains a video camera and even three differential optical absorption spectroscopy (DOAS) telescopes looking down, above, and below the horizon. This adds a remote-sensing capability to CARIBIC.

Figure 2 is a frontal photograph of the CARIBIC phase 2 measurement container. The container weighs ~ 1450 kg and is 3.2 m wide, 1.6 m deep, and 1.5 m high. Equipment is fixed in seven racks mounted on shock absorbers on the double-floor system. Two power supplies convert most of the required aircraft power (peak load 7 kW) to 28 V dc. One computer controls the 18 devices, which are all fitted with individual control and data-acquisition systems. The container with its smoke detection and ventilation systems has been extensively tested for possible electromagnetic interferences.

Table 1 lists the atmospheric species that can be measured or collected by CARIBIC phase 2 and the detection principle used. Compared with phase 1, notably the PTRMS instrument (Proton Transfer Reaction Mass Spectrometer), the gaseous water detection, the fast CO analyzer, the absorption tube system for volatile organic carbon, the 28 sample capacity glass (inert) bottle sampler, the DOAS based remote sensing system, and the optical particle counter for climate relevant aerosol sizes, signify a major increase in the analytical capabilities of CARIBIC.

Scientific Results

The main information obtained by airliner projects like CARIBIC pertains to the atmosphere at generally 9–11 km, except for Boeing 747 aircraft, which have cruising altitudes up to 12 km. Generally at mid-latitudes and in the subtropics, stratospheric air is regularly intercepted; in the tropics, upper to middle tropospheric air is probed. With long-range flights into the Southern Hemisphere, valuable data from this hemisphere and for the tropics including the Intertropical Convergence Zone (ITCZ) are obtained.

The information that CARIBIC phase 1 delivered has been partly published (see www.caribic-atmospheric.com). The systematic, simultaneous measurements of CO and O_3 have allowed the assessment of the upper tropospheric budgets of CO and O_3 using CO-O_3 correlations over various distances [*Zahn et al.*, 2002]. Measurement of the contrasting behavior of CO and O_3 across the tropopause has furthermore allowed the defining of an extratropical chemical tropopause [*Zahn et al.*, 2004a] and the better quantifying of the ventilation of the lowermost stratosphere [*Zahn et al.*, 2004b].

The analyses of fine aerosol particles have led to a first climatology [*Hermann et al.*, 2003] of these short-lived compounds. Biomass burning and convection over Africa have been studied using nonmethane hydrocarbon (NMHC) analyses [*Mühle et al.*, 2002], and high aerosol abundances were reported [*Heintzenberg et al.*, 2003]. The sulphur loading of the tropopause region by fine aerosol particles has also been investigated [*Martinsson et al.*, 2001].

The combined measurement of NO and NO_y have allowed the determination of the emission ratio of condensation nuclei due to aircraft emissions. The value obtained agrees well with the average value obtained during the SONEX (Subsonic Assessment of Ozone and Nitrogen Oxides) campaign using the NASA DC-8 research aircraft (unpublished results).

CARIBIC phase 1 work in progress deals with the gradient of cosmogenic ^{14}CO (carbon-14 monoxide) across the tropopause, the summer monsoon plume from SE Asia, the composition and ozone formation in the North American plume over the Atlantic, sources of NMHCs, and a reassessment of the global atmospheric budget of molecular hydrogen using D/H isotope analysis.

The CARIBIC phase 2 system with an Airbus A340-600 became operational in December 2004. Figure 3 depicts the range of this aircraft from Frankfurt, Germany. Lufthansa Airlines allocates certain flight slots based on cargo requirements and suitable long-distance destinations, upon which optimal flight possibilities are selected by the CARIBIC science panel. Before flight, the container and equipment are tested and calibrated, transported from the Max Planck Institute for Chemistry to Frankfurt Airport, thoroughly inspected and tested again, and loaded into the forward cargo compartment. After locking the container into position, and connecting air tubing and electrical and data systems, a vital function system test is performed. Apart from smoke detection and a master switch under operation of the pilot, the entire process of sequential switching on and measuring is automated.

Upon return from the back-to-back intercontinental flights (outbound and return flight), the container is unloaded and transported

Fig. 3. The range from Frankfurt, Germany, of the Airbus A360-600.

Table 1. Phase 2 Instrumentation for Trace Gas (plus O_2) and Aerosol Particle Analyses.

		Trace Compound	*Instrument*
1	#	O_3 ultra-fast	Chemi-luminescence on an organic dye
2	#	O_3 accurate and precise	UV Photometer
3	#	CO	Vacuum UV-fluorescence
4		H_2O gas phase	Photo-acoustic detector with diode laser
5	#	H_2O total	Chilled mirror and photo-acoustic detector
6	#	NO	Chemi-luminescence reaction with O_3
7	#	NOy	Chemi-luminescence (conversion to NO)
8		Hg	Enrichment and atomic fluorescence
9		CO_2	NDIR
10		O_2 ultra high precision, 10 per meg	Electrochemical cells
11		Methanol, Acetone, Acetaldehyde	Proton Transf. React. Mass-Spec. (PTR-MS)
12	#	Aerosol with diameter >4 nm	Condensation Particle Counter (CPC)
13	#	Aerosol with diameter >12 nm	Condensation Particle Counter (CPC)
14	#	Aerosol with diameter >18 nm	Condensation Particle Counter (CPC)
15		Aerosol size distribution 150-5000 nm	Optical Particle Counter (OPC)
16	#	Aerosol elemental composition	Impactor, and analysis with PIXE
17		Particle morphology	Impactor, and electron microscopy
18		VOC samples	Absorption tubes and GC-MS analysis
19	#	NMHC, halogens, greenhouse gases	Sampling in glass bottles and GC analysis
20		BrO, HCHO, OClO, O_4	DOAS
21		Cirrus (under certain conditions)	Camera
22		Physical data	Aircraft ARINC system

Pound sign indicates CARIBIC phase 1 capabilities. For more information about equipment, institutions, and PI's, the reader is referred to the CARIBIC Web site.

back to Mainz, near Frankfurt Airport. Here the data retrieval begins, followed by the extensive laboratory analysis of the air and aerosol samples. Because Lufthansa (Star Alliance) equips all its long-range aircraft with a broadband-based wireless Local Area Network (LAN) facility, possibilities for real-time data retrieval in flight are possible in the future.

Outlook

Experience gained during phase 1 of CARIBIC has provided confidence about the technical and logistical operational soundness of the container concept. Given the similarity between phase 1 and 2, a smooth and regular operation is expected. In view of the extensive list of equipment in Table 1 and considering the results obtained during phase 1, it may be concluded that valuable new insights into the chemistry and composition of the atmosphere and their changes will result from CARIBIC. The addition of the mercury and highly precise O_2 measurements are examples. The operation of a DOAS system, the PTR mass spectrometer, the expected data for gaseous and total water, and the extensive aerosol data add to the overall value of the data sets. It is expected that in view of the increasing sophistication of atmospheric models, particularly detailed data set will have great value in the future.

It is hoped that CARIBIC phase 2 will be operational over a long time period in order to obtain an optimally consistent set of data. It is obvious that certain flight routes are most useful, especially those covering the largest range of latitudes. At least monthly flights on those strategic routes are needed to obtain representative information on seasonal changes. Embedded in those regular flights, occasionally other destinations can be targeted for exploring other regions of the global atmosphere. For instance, during phase 1, three regions were well-characterized, namely, Europe to the Indian Ocean, Europe to the Caribbean, and parts of Africa. Although aircraft exhaust plumes have been detected, generally the movement of air masses relative to the air traffic corridors is such that basically background air is probed.

Two related questions one may ask are; what exactly is the optimal flight frequency, and is there a need for more aircraft of the CARIBIC type? Phase 1 showed that our institutions could handle at the most one to two flights per month, but there is no fundamental reason why this number cannot be increased. Provided the airline can offer a sufficient variety of destinations within the operational constraints of the aircraft, such would still add value to the overall CARIBIC operation. A limiting factor remains, however, that certain important and undoubtedly interesting parts of the globe cannot be covered solely by flying from Europe. In this respect it is desirable to have flights not only from Japan (JAL) and Europe but also from North America.

One task is the integration of the CARIBIC data with existing observational networks. The World Meteorological Organization strongly supports and promotes the Global Atmophere Watch program, and there is a strong need to forge a coherent global observational capacity.

The existing CARIBIC flights already have added upper and middle tropospheric data for the greenhouse gases and CO to the extensive invaluable inventory of the NOAA/CMDL and other network data for trace gases. CARIBIC measurements also can assist in the validation of certain satellite-based measurements, and the AURA and ENVISAT observations may benefit from this. It is also clear that in this respect, the 10-year time horizon of CARIBIC is of importance in achieving continuity and consistency for a large range of data.

Acknowledgments

The development and operation of CARIBIC has been financially supported by the German Ministry of Education and Science (AFO 2000), and by the European Commission, DGXII Environment RTD, 4th and 5th Framework program. We acknowledge Lufthansa Airlines and Lufthansa Technik for their support of CARIBIC.

References

Brenninkmeijer, C. A. M., et al. (1999), CARIBIC—Civil aircraft for global measurement of trace gases and aerosols in the tropopause region, *J. Atmos. Oceanic Technol., 16*, 1373–1383.

Falconer, P. D., and J. D. Holdeman (1976), Measurements of atmospheric ozone made from a GASP-equipped 747 airliner: Mid-March, 1975, *Geophys. Res. Lett., 3*, 101–104.

Heintzenberg, J., M. Hermann, and D. Theiss (2003), Out of Africa: High aerosol concentrations in the upper troposphere over Africa, *Atmos. Chem. Phys., 3*, 1191–1198.

Hermann, M., J. Heintzenberg, A. Wiedensohler, A. Zahn, G. Heinrich, and C. A. M. Brenninkmeijer (2003), Meridional distributions of aerosol particle number concentrations in the upper troposphere and lower stratosphere obtained by Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container (CARIBIC) flights, *J. Geophys. Res., 108*, 4114, doi:10.1029/2001JD001077.

Martinsson, B. G., G. Papaspiropoulos, J. Heintzenberg, and M. Hermann (2001), Fine mode particulate sulphur in the tropopause region measured from intercontinental flights (CARIBIC), *Geophys. Res. Lett., 28*, 1175–1178.

Mühle, J., C. A. M. Brenninkmeijer, T. S. Rhee, F. Slemr, D. E. Oram, S. A. Penkett, and A. Zahn (2002), Biomass burning and fossil fuel signatures in the upper troposphere observed during a CARIBIC flight from Namibia to Germany, *Geophys. Res. Lett., 29*, 1910, doi:10.1029/2002GL015764.

Pratt, R., and P. Falconer (1979), Circumpolar measurements of ozone, particles, and carbon monoxide from a commercial airliner, *J. Geophys. Res., 84*, 7876–7882.

Zahn, A., C. A. M. Brenninkmeijer, W. A. H. Asman, P. J. Crutzen, G. Heinrich, H. Fischer, J. W. M. Cuijpers, and P. F. J. van Velthoven (2002), The budgets of O_3 and CO in the upper troposphere: CARIBIC passenger aircraft results 1997-2001, *J. Geophys. Res., 107*, 4337, doi:10.1029/2001JD001529.

Zahn, A., C. A. M. Brenninkmeijer, and P. F. J. van Velthoven (2004a), Passenger aircraft project CARIBIC 1997-2002: Part I. The extratropical chemical tropopause, *Atmos. Chem. Phys. Discuss., 4*, 1091–1117.

Zahn, A., C. A. M. Brenninkmeijer, and P. F. J. van Velthoven (2004b), Passenger aircraft project CARIBIC 1997-2002: Part II. The ventilation of the lowermost stratosphere, *Atmos. Chem. Phys. Discuss., 4*, 1119–1150.

Author Information

C. A. M. Brenninkmeijer, F. Slemr, C. Koeppel, D. S. Scharffe, M. Pupek, J. Lelieveld and P. Crutzen, Max Planck Institute for Chemistry, Mainz, Germany; A. Zahn, D. Sprung, and H. Fischer, Institute for Meteorology and Climate Research, Karlsruhe, Germany; M. Hermann, M. Reichelt, and J. Heintzenberg, Institute for Tropospheric Research, Leipzig, Germany; H. Schlager, H. Ziereis, and U. Schumann, Institute for Atmospheric Physics, DLR, Oberpfaffenhofen, Germany; B. Dix and U. Platt, University of Heidelberg, Germany; R. Ebinghaus, GKSS, Geesthacht, Germany; B. Martinsson, University of Lund, Sweden; P. Ciais and D. Filippi, LSCE, CEA, CNRS, Paris, France; M. Leuenberger, University of Bern, Switzerland; D. Oram and S. Penkett, University of East Anglia, Norwich, U.K.; P. van Velthoven, Royal Dutch Meteorological Organization, de Bilt, the Netherlands; and A. Waibel, German Lufthansa, Frankfurt Airport, Germany

For additional information, contact C. A. M. Brenninkmeijer; E-mail carlb@mpch-mainz.mpg.de. For complete project information, see www.caribic-atmospheric.com.